A Sponge-based Moving Bed Bioreactor for Micropollutant Removal from Municipal Wastewater

by

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CERTIFICATE OF ORIGINAL AUTHORSHIP

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

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List of Abbreviations

AOP	Advanced oxidation process
BAC	Biological activated carbon
CAFO	Concentrated animal feeding operation
COD	Chemical oxygen demand
CAS	Conventional activate sludge
DBP	Di-butyl phthalate
DEET	N,N-diethyl-meta-toluamide
DEHP	Di(2-ethylhexyl) phthalate
DMP	Di-methyl phthalate
DO	Dissolved oxygen
DOM	Dissolved organic matter
EDC	Endocrine disrupting compound
EPS	Extracellular polymeric substance
GAC	Granule activated carbon
GC-MS	Gas chromatography-mass spectrometry
HRT	Hydraulic retention time
MBBR	Moving bed bioreactor
MBR	Membrane bioreactor
MB-SMBR	Moving bed-submerged membrane bioreactor
MF	Microfiltration
MLSS	Mixed liquor suspended solids
MLVSS	Mixed liquor volatile suspended solids
NF	Nanofiltration
NLR	Nitrogen loading rate
NOM	Natural organic matter
NSAID	Nonsteroidal anti-inflammatory drug
OLR	Organic loading rate
PAC	Powdered activated carbon
PCP	Personal care product
PNEC	Predicted no effect concentration
PPCP	Pharmaceutical and personal care product

PU	Polyurethane
PVA	Polyvinyl alcohol
PVDF	Polyvinylidene fluoride
RO	Reverse osmosis
SAnMBR	Submerged anaerobic membrane bioreactor
SBBGR	Sequencing batch biofilter granular reactor
SBF	Sponge biofilter
SBR	Sponge batch reactor
SMBR	Submerged membrane bioreactor
SMP	Soluble microbial products
SND	Simultaneous nitrification–denitrification
SPE	Solid phase extraction
SRT	Sludge retention time
TCEP	Tris(2-chloroethyl) phosphate
TCPP	Tris(1-chloro-2-propyl) phosphate
TMP	Transmembrane pressure
TN	Total nitrogen
TOC	Total organic carbon
TP	Total phosphorus
UF	Ultrafiltration
WWTP	Wastewater treatment plant

List of Symbols

Cs, a	Concentration of micropollutant on the attached biosolids (μg/g)
Cs, s	Concentration of micropollutant on the suspended biosolids (μg/g)
$C_{w,\ eff}$	Average effluent concentrations of micropollutants (ng/L)
$C_{w,\ inf}$	Average influent concentrations of micropollutants (ng/L)
J	Permeation flux ($L/m^2 \cdot h$)
K_d/D	Solid-water distribution coefficient
K_H	Henry's law constant
K_{OW}	Octanol–water partition coefficient
L_{sol}	Load of micropollutant removed via sorption over 30 days (ng)
MLSS	Mixed liquor suspended biosolids concentration (g/L)
pK_a	Acid dissociation constant
ΔP_T	Transmembrane pressure (kPa)
Q	Flow rate of the MBBR (L/day)
R_c	Cake resistance formed by cake layer deposited over membrane surface (m^{-1})
R_f	Fouling resistance caused by pore plugging and/or solute adsorption onto the membrane pore and surface (m^{-1})
R_m	Intrinsic membrane resistance caused by membrane itself and permanent resistance (m^{-1})
ΔSS	Increased amount of attached biosolids over the study period (g)
T	Duration of the study period (day)
μ	Viscosity of the permeate (m^2/s)

Abstract

Over the past few decades, the frequent detection of micropollutants in the aquatic environment has raised particular health and environmental concerns. Wastewater treatment plants (WWTPs) serve as significant barriers to reduce the release of micropollutants. However, due to the diverse characteristics and low concentrations of micropollutants, WWTPs can only achieve variable and often inadequate removals, ranging from 12.5% to 100% for some frequently reported compounds.

This study investigated a sponge-based MBBR for its effectiveness in the elimination of various micropollutants, including pharmaceuticals and personal care products, steroid hormones, industrial chemicals and pesticides. A moving bed-submerged membrane bioreactor (MB-SMBR) was also evaluated in terms of micropollutant removal and membrane fouling.

During the batch experiments, non-acclimatized (virgin) sponge showed significant and rapid sorption of hydrophobic compounds. Acclimatized sponge could achieve much higher elimination of some acidic pharmaceutical compounds, such as acetaminophen, diclofenac, gemfibrozil, ibuprofen, ketoprofen, naproxen and salicylic acid. Carbamazepine, fenoprop and metronidazole were poorly removed during all the batch experiments.

The sponge-based MBBR was effective in removing organics and nutrients (except PO_4-P). Most of the selected micropollutants (16 out of 22) showed removals of higher than 70%. The poorly or moderately removed compounds included carbamazepine (25.9%), fenoprop (31.0%), diclofenac (45.7%), metronidazole (54.8%), ketoprofen (58.2%), and gemifibrozil (62.4%). The low biodegradability and/or polar property were two causes for the insufficient elimination. Overall, the effectiveness of the MBBR for micropollutant removal was comparable with those of other biological treatment processes, including activated sludge and membrane bioreactors (MBRs). Biodegradation was the major removal mechanisms for most compounds during the MBBR treatment. Sorption was only significant for the

refractory compounds, while the readily biodegradable compounds did not considerably accumulate on the biosolids.

With the incorporation of the SMBR, the whole system (MB-SMBR) could significantly reduce the effluent turbidity. However, the SMBR did not achieve much supplementary removal of organic, nutrients and micropollutants. The membrane fouling in the SMBR occurred to a minor extent during the first 84 days of operation, after which an abrupt TMP increase was observed. High EPS levels (16.24 mg/L) in the SMBR was a potential cause for the severe fouling. The overgrowth of filamentous bacteria could also been deemed a factor that accelerated the membrane fouling rate. The total membrane resistance was mainly attributed to the deposited cake layer (76.5%), followed by the pore blocking (12.0%), clean membrane resistance (10.5%) and irreversible fouling (1.0%).